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Linear Response and Stochastic Resonance of Superparamagnets

T. F. Ricci[1] **and C. Scherer**[1]

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The standard mathematical expression for the response functions of the linear response theory for Markovian stochastic systems, constructed from the corresponding Fokker–Planck equation, is transformed into an expression which is very suitable for numerical simulations. The method is applied to a stochastic model for superparamagnetism presented previously by the authors. For convenient values of the parameters the model shows the phenomenon of stochastic resonance.

KEY WORDS: Stochastic processes; linear response; superparamagnetism; monodomain, stochastic resonance.

1. LINEAR RESPONSE OF STOCHASTIC SYSTEMS

A superparamagnetic particle is an example of a mesoscopic physical system whose states are described by a set of variables which are random functions of time, or stochastic processes (SP), which may be represented by an M-dimensional vector $\mathbf{X}(t) = (X_1(t), X_2(t),..., X_M(t))$. In the present work we restrict the treatment to mesoscopic systems for which the $X_i(t)$ are Markovian SP described by a Langevin type of stochastic differential equation,[2]

$$dX_i(t) = A_i(\mathbf{X}(t), t)\, dt + \sum_{j=1}^{M} B_{ij}(\mathbf{X}(t), t)\, dW_j(t) \tag{1}$$

where the coefficients A_i and B_{ij} are functions of the random variables and the time. The $dW_j(t) = W_j(t+dt) - W_j(t)$ are the infinitesimal increments

[1] Instituto de Fisica, Universidade Federal do Rio Grande do Sul, 91501-970 Porto Alegre, RS, Brazil.

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of the orthogonal, normalized, Wiener processes $W_j(t)$ (the intensity of the noise is included in the B_{ij} coefficients). Therefore, $\langle dW_i(t)\rangle = 0$ and $\langle dW_i(t)\, dW_j(t)\rangle = \delta_{ij}\, dt$, where $\langle \cdots \rangle$ means average over all realizations of the Wiener process.

As is well known (e.g., ref. 2), the probability density $P(\mathbf{x}, t)$ for the SP $X_i(t)$ described by Eq. (1) obeys the following Fokker–Planck equation (FPE),

$$\frac{\partial}{\partial t} P(\mathbf{x}, t) = \hat{L}(t)\, P(\mathbf{x}, t) \tag{2}$$

where the "Fokker–Planck operator" $\hat{L}$ is given by ($\partial_i = \partial/\partial x_i$)

$$\hat{L}(t) = -\sum_{i=1}^{M} \partial_i A_i(\mathbf{x}, t) + \frac{1}{2} \sum_{i,j=1}^{N} \partial_i \partial_j D_{ij}(\mathbf{x}, t) \tag{3}$$

and

$$D_{ij} = \sum_{k=1}^{M} B_{ik} B_{jk}$$

are the elements of the diffusion matrix. The transition probability density $P(\mathbf{x}, t \mid \mathbf{x}', t')$, also satisfies Eq. (2), with the special initial condition $P(\mathbf{x}, t' \mid \mathbf{x}', t') = \delta(\mathbf{x} - \mathbf{x})$.

In the next section we will study the response of a system of superparamagnetic particles to a weak, oscillating applied field. For this reason we review now briefly the essential results of linear response theory and transform the standard result for the response functions[3] into a form more appropriate for numerical simulations.

Consider the class of stochastic systems whose associated FPE satisfies the following conditions:

(i) The diffusion coefficients are not explicitly time dependent, i.e., $D_{ij} = D_{ij}(\mathbf{x})$.

(ii) The drift coefficients can be separated into a term linear in the applied fields $F_j(t)$ and another that, if present, is independent of the F_j, i.e.,

$$A_i = \sum_{j=1}^{M} \gamma_{ij}(\mathbf{x})\, F_j(t) + A_i^{(0)}(\mathbf{x}) \tag{4}$$

These requirements are not too restrictive and this class of FPEs is vast enough to comprise a large number of phenomena in classical nonequilibrium statistical physics.[2,3] In particular, requirement (ii) is always satisfied in the limit of infinitesimal applied fields (linear response limit). If a static field $F_j^{(0)}$ is present it can be considered part of the system and its effect included in the $A_i^{(0)}(\mathbf{x})$.

The system is assumed in equilibrium with the static field $F_j^{(0)}$ in the remote past, $t = -\infty$, so that its initial probability density $P_{st}^{(0)}(\mathbf{x})$ satisfies the unperturbed FPE

$$\hat{L}_0 P_{st}^{(0)}(\mathbf{x}) = 0 \tag{5}$$

with

$$\hat{L}_0 = -\sum_{i=1}^{M} \partial_i A_i^{(0)}(\mathbf{x}) + \sum_{i,j=1}^{M} \partial_i \partial_j D_{ij}(\mathbf{x}) \tag{6}$$

The conditional probability density $P^{(0)}(\mathbf{x}, t \mid \mathbf{x}^p, 0)$, in the absence of perturbation and given the initial value $\mathbf{X}(0) = \mathbf{x}^p = (x_1^p, x_2^p, ..., x_M^p)$, satisfies the FPE Eq. (2) with $\hat{L}_0$ in place of $\hat{L}(t)$, and formal solution

$$P^{(0)}(\mathbf{x}, t \mid \mathbf{x}^p, 0) = \exp[t\hat{L}_0]\, \delta(\mathbf{x} - \mathbf{x}^p) \tag{7}$$

Denoting by $\langle X_i(t \mid \mathbf{x}^p) \rangle_0$ the expectation value of $X_i(t)$ in the absence of perturbation and given that $\mathbf{X}(0) = \mathbf{x}^p$, the above equation implies

$$\langle X_i(t \mid \mathbf{x}^p) \rangle_0 = \int x_i \exp[t\hat{L}_0]\, \delta(\mathbf{x} - \mathbf{x}^p)\, d^M x = \exp[\hat{L}_0^\dagger]\, x_i^p \tag{8}$$

where we have used the identity

$$\int f(\mathbf{x}) \exp[t\hat{L}_0]\, g(\mathbf{x})\, d^M x \equiv \int g(\mathbf{x}) \exp[t\hat{L}_0^\dagger]\, f(\mathbf{x})\, d_x^M \tag{9}$$

which defines the adjoint operator $\hat{L}_0^\dagger$ of $\hat{L}$,

$$\hat{L}_0^\dagger = +\sum_{j=1}^{M} A_i^{(0)}(\mathbf{x})\, \partial_i + \sum_{i,j=1}^{M} D_{ij}(\mathbf{x})\, \partial_i \partial_j \tag{10}$$

Equation (8) tells us that $\exp[t\hat{L}_0^\dagger]$ may be interpreted as the *envolution operator for the expectation value of* $\mathbf{X}(t)$.

When a time-dependent perturbing field $F_j(t)$ is applied, the change in the expectation value of $X_i(t)$, with respect to its unperturbed value, in the linear response limit; is

$$\langle X_i(t) \rangle - \langle X_i \rangle_0 = \sum_{j=1}^{M} \int_{-\infty}^{t} \Phi_{ij}(t - t')\, F_j(t')\, dt' \tag{11}$$

where the response functions $\Phi_{ij}(t-t')$ are given by[3]

$$\Phi_{ij}(t-t') = -\sum_{k=1}^{M} \int x_i \exp(t-t')\, \hat{L}_0\, \partial_k[\gamma_{kj}(\mathbf{x})\, P_{st}^{(0)}(\mathbf{x})]\, d^M x \qquad (12)$$

This standard formal expression for $\Phi_{ij}(t)$ is not appropriate for numerical calculations. Moreover, it is usually very difficult to solve the unperturbed FPE to obtain the probability density $P_{st}^{(0)}(x)$. Some limit cases for which the integration in Eq. (12) may be performed exactly, will be seen in the next section. For $M \geqslant 2$ a general analytical form for $P_{st}^{(0)}(\mathbf{x})$ is available only if the unperturbed FPE satisfies detailed balance conditions (DBC).[4] For this case Graham[5] presents an expression for the response functions in the form of expectation values which may be used for numerical simulations. A more convenient expression which is both simpler and more generals because it does not depend on DBC may be derived as follows.

Using Eq. (9) and performing an integration by parts, one can transform Eq. 12 into

$$\Phi_{ij}(t-t') = \sum_{k=1}^{M} \int P_{st}^{(0)}(\mathbf{x})\, \gamma_{kj}(\mathbf{x})\, \partial_k\{\exp[(t-t')\, \hat{L}_0^{\dagger}]\, x_i\}\, d^M x \qquad (13)$$

Equation (8) may be used in Eq. (13) to transform it further into the expression

$$\Phi_{ij}(t) = \sum_{k=1}^{M} \langle \gamma_{kj}(\mathbf{x})\, \partial_k \langle X_i(t\,|\,\mathbf{x})\rangle_0 \rangle_{st} \qquad (14)$$

where the symbol $\langle \cdots \rangle_{st}$ indicates the average over the initial point $\mathbf{x}$ distributed according to the unperturbed stationary probability density $P_{st}^{(0)}(\mathbf{x})$. In particular, $\Phi_{ij}(0) = \langle \gamma_{ij}(\mathbf{x})\rangle_{st}$. As we will see in the next section, Eq. (14) is very suitable for calculating the response functions by numerical simulations based on the Langevin equation (1), without the need of knowing $P_{st}^{(0)}(\mathbf{x})$.

2. APPLICATION TO SUPERPARAMAGNETS

A superparamagnet is a material containing very fine ferromagnetic particles, whose volumes are sufficiently small for each particle to be a magnetic monodomain having, therefore, a magnetic moment $\boldsymbol{\mu}$. The concentration of magnetic particles in the material is sufficiently low for the interaction between them to be negligible. We define the *spin* of a particle by $\mathbf{S} \equiv \boldsymbol{\mu}/\gamma_e$, where γ_e is the electronic gyromagnetic ratio.

The interaction energy of the particle's spin with the environment is $V(\mathbf{S}, t) = V_{ext} + V_A + V_D$, where V_{ext} is the interaction the applied field, V_A is the magnetocrystalline energy due to the crystal anisotropy, and V_D is the magnetostatic energy corresponding to the interaction of $\mathbf{S}$ with the demagnetization field. It is convenient to define an *effective magnetic field* by $\mathbf{H}_{eff} = -\partial V/\partial \mathbf{S}$ (we take $\gamma_e = 1$ for simplicity). The system also interacts with the thermal excitations of the crystalline lattice: phonons produce movements of electrical charges in the particle and these very rapidly fluctuating electrical currents exert torques on the spin $\mathbf{S}$. We do not include this fluctuating interfction in V. Instead, we will treat it as a *white noise torque* $\mathbf{\Gamma}(t)$. The movement of $\mathbf{S}(t)$ under the influence of the lattice and effective field is known as *Néel relaxation.*[6] A stochastic differential equation of the Langevin type may then be written for $\mathbf{S}$,

$$\frac{d\mathbf{S}}{dt} = \mathbf{\Omega}(\mathbf{S}, t) + \mathbf{\Gamma}(t) \tag{15}$$

where $\mathbf{\Omega}$ represents the deterministic part of the torque. In a previous paper on the subject[1] we derived an equation of motion for a "classical spin" in the presence of relaxation and noise from a generalized Lagrangian formalism. In the limit of zero noise our result reproduces the Landau–Lifshitz equation for $\mathbf{S}(t)$,[7]

$$\frac{d\mathbf{S}}{dt} = \gamma(S^2)\left[\mathbf{S}\times\mathbf{H}_{eff} - \frac{\lambda}{S^2}\mathbf{S}\times(\mathbf{S}\times\mathbf{H}_{eff})\right] \tag{16}$$

where λ is the relaxation constant, S is the magnitude of $\mathbf{S}$, and $\gamma(S^2) = S^2/(\lambda^2 + S^2)$. In this equation, S is a constant of motion, which is a good approximation for not too small superparamagnetic particles. However, if the number of atomic spins making up $\mathbf{S}$ is not very large and the temperature is not too low, the fluctuations of S caused by the interaction of the atomic spins with the random currents may become important. A term of the form $\Psi(S)\,\hat{e}_3$, where $\hat{e}_3$ is the unit vector in the direction of $\mathbf{S}$, allows for changes in S. Assuming that these changes are small fluctuations around a most probable value S_0, we keep for Ψ only a linear term, i.e., $\Psi - \beta(S - S_0)$, where β is called the *longitudinal relaxation constant.* With this generalization, the magmtude of $\mathbf{S}$ in the Landau–Lifshitz equation will always relax to S_0. The vector $\mathbf{\Omega}$ in Eq. (15) is accordingly written as

$$\mathbf{\Omega}(S, t) = \gamma(S^2)\left[\mathbf{S}\times\mathbf{H}_{eff} - \frac{\lambda}{S^2}\mathbf{S}\times\mathbf{H}_{eff})\right] - \beta(S - S_0)\,\hat{e}_3 \tag{17}$$

The characteristic time scale for the movement of $\mathbf{S}$ is $\sim 10^{-10}$ sec,[8] while the characteristic time scale for the random torque $\Gamma(t)$ is the inverse frequency of the optical phonons, i.e., $\sim 10^{-13}$ sec. Considering, moreover, that the random torque is the sum of the contributions of many independent phonons, we are led to treat it as Gaussian white noise. Therefore we need to specify only its first and second moments to define whole statistics. Assuming its Cartesian components to be statistically independent and isotropic, we may write $\langle \Gamma_i(t) \rangle = 0$ and $\langle \Gamma_i(t)\, \Gamma_j(t') \rangle = 2D\, \delta_{ij}(t-t')$, where $i, j = x, y, z$ and D is the noise intensity.

To apply the results of Section 2 on our superparamagnet, we write Eq. (15) in the form of Eq. (1). Defining the vector $\mathbf{X} = \mathbf{S}(t)/S_0$, whose modulus will be denoted by R, and writing the effective field as $\mathbf{H}_{\text{eff}} = \mathbf{H}^{(0)}(\mathbf{X}) + \delta\mathbf{H}(t)$, where $\delta\mathbf{H}(t)$ is the perturbing field [$F_j(t)$ in the notation of Eq. (4)], we come to the following identifications:

$$A_i^{(0)}(\mathbf{X}) = \beta \left(\frac{1-R}{R} \right) X_i + \sum_{j=1}^{3} \gamma_{ij}(\mathbf{X})\, H_j^{(0)}(\mathbf{X}) \tag{18}$$

$$\gamma_{ij}(\mathbf{X}) = -\gamma(R^2) \left[\sum_{k=1}^{3} \varepsilon_{ijk} X_k + \frac{\lambda}{S_0 R^2} (X_i X_j - \delta_{ij} R^2) \right] \tag{19}$$

where ε_{ijk} is the Levi–Civita antisymmetric tensor and

$$\gamma(R^2) = \frac{R^2}{R^2 + (\lambda/S_0)^2}$$

$$B_{ij} = \delta_{ij} \left(\frac{2D}{S_0^2} \right)^{1/2} \tag{20}$$

$$dW_j(t) = \frac{1}{\sqrt{2D}} \int_t^{t+dt} \Gamma_j(t')\, dt' \tag{21}$$

In the limit of no fluctuations of R, i.e., for $R(t) = 1$, this model reduces to that proposed by Brown.[9] To see which conditions our parameters have to satisfy for this limit to be achieved, we work with the longitudinal component of Eq. (15), namely

$$dR(t) = -\beta(R-1)\, dt + \left(\frac{2D}{S_0^2} \right)^{1/2} dW_3(t) \tag{22}$$

where $dW_3(t) = \hat{e}_3 \cdot d\mathbf{W}(t)$ is the projection of the noise pin the direction of $\mathbf{X}$. Using the relation

$$\hat{e}_3(t) = \hat{\imath} \sin\theta \cos\phi + \hat{\jmath} \sin\theta \sin\phi + \hat{k} \cos\theta$$

we find

$$dW_3(t) = \sin\theta \cos\phi \, dW_x(t) + \sin\theta \sin\phi \, dW_y(t) + \cos\theta \, dW_z(t)$$

Therefore $R(t)$ satisfies a stochastic differential equation with "multiplicative noise." The transformation of coordinates was carried out using the rules of ordinary calculus, which means that Eq. (22) is to be understood as a Stratonovich–Langevin equation and therefore $\langle dW_3(t)\rangle \neq 0$. The corresponding Ito–Langevin equation is[2]

$$dR(t) = -\beta(R-1) + \frac{2D}{S_0^2 R} + \left(\frac{2D}{S_0^2}\right)^{1/2} dW_3(t)$$

for which $\langle dW_3(t)\rangle = 0$. This equation does not depend on θ or ϕ, and the corresponding one-dimensional FPE has the following stationary solution:

$$G_{st}(r) = N_0 r^2 \exp[-(r-1)^2/2\sigma^2]$$

where $\sigma^2 \equiv D/\beta S_0^2$. Therefore this model reduces to Brown's model when $D \ll \beta S_0^2$.

We consider two cases of static field $\mathbf{H}^{(0)}(\mathbf{X})$:

Case I. A static, uniform magnetic field is applied along the z direction and no other forces act on $\boldsymbol{\mu}$. The interaction energy between the applied field $\mathbf{H}^{(0)}$ in the z direction and the magnetic moment $\boldsymbol{\mu}$ is $V^{(0)} - \mu_z H^{(0)}$.

Case II. There is a crystalline effective field derived from the magnetic crystalline anisotropy energy $V^{(0)}(\mathbf{x}) = Kv \sin^2\theta = Kv(x^2 + y^2)/r^2$, where θ is the angle between $\boldsymbol{\mu}(t)$ and the easy axis (the z-axis), and K is the anisotropy constant. Then

$$H_x^{(0)} = -\frac{2Kv}{r^4} xz^2$$

$$H_y^{(0)} = -\frac{2Kv}{r^4} yz^2$$

$$H_z^{(0)} = +\frac{2Kv}{r^4} z(x^2 + y^2)$$

and no other torques act on $\boldsymbol{\mu}$.

In appendix A we show that for these cases the model does not satisfy DBC.

2.1. Zero-Noise Limit

It is interesting to analyze Eq. (14) in the limit of zero intensity of the noise, $D \to 0$.

For both cases considered above we conclude straightforwardly by invoking the Langevin equation Eq. (1) that in this limit:

(i) the magnitude of $\mathbf{X}(t)$ will be a constant, $r = 1$, so that the phase points will move over a unitary sphere.

(ii) All realizations of $\mathbf{X}(t \mid \mathbf{x}^p)$ converge in mean square (see, e.g., ref. 10) to the deterministic trajectory of the system, i.e.,

$$\lim_{D \to 0} [X_i(t \mid \mathbf{x}^p) - x_i(t \mid \mathbf{x}^p)]^2 = 0$$

where the function $x_i(t \mid \mathbf{x}^p)$ is the solution of Eq. (1) for $B_{ij} = 0$, $A_i = A_i^{(0)}$, and initial condition $\mathbf{X}(0) = \mathbf{x}^p = (x_p, y_p, z_p)$. In both cases considered here we have obtained the deterministic trajectories (see Appendix B) and therefore $\langle X_i(t \mid \mathbf{x}^p) \rangle_0$.

(iii) In the absence of noise all solutions of the Langevin equation relax to a minimum-energy state; therefore the equilibrium probability density $P_{st}^{(0)}(\mathbf{x}^p)$ will be the products of Dirac delta functions centered on the energy minima.

The response function Eq. (14) and the susceptibility Eq. (25) will be considered separately for the two cases in the limit $D \to 0$.

Case I. The only energy minimum is for $\mathbf{x} = (0, 0, 1)$ and therefore $P_{st}^{(0)}(\mathbf{x}^p) = \delta(x_p)\, \delta(y_p)\, \delta(z_p - 1)$. Using this distribution, identifying $\langle X_i(t \mid \mathbf{x}^p) \rangle_0$ with the deterministic trajectory given in Appendix B, and using Eq. (19) for the γ_{ij} we find that Eq. (14) is readily integrable, leading to

$$\Phi_{11}(t) = \Phi_{22}(t) = \frac{1}{\sqrt{1 + (\lambda/S_0)^2}}\, e^{-t/\tau} \sin(\omega_0 t + \delta_0)\, u(t) \tag{23}$$

$$\Phi_{12}(t) = -\Phi_{21}(t) = -\frac{1}{\sqrt{1 + (\lambda/S_0)^2}}\, e^{-t/\tau} \cos(\omega_0 t + \delta_0)\, u(t) \tag{24}$$

and $\Phi_{13} = \Phi_{23} = \Phi_{31} = \Phi_{32} = \Phi_{33} = 0$ where $\tau = (S_0^2 + \lambda^2)/\lambda S_0 H^{(0)}$, $\omega_0 = S_0/\lambda\tau$, and $\delta_0 = \arctan[\lambda/S_0]$.

The complex admittances or susceptibility, defined as the Fourier–Laplace transform of $\Phi_{ij}(t)$,

$$\chi_{ij}(\omega) = \int_0^{\infty} \Phi_{ij}(t) \exp(i\omega t)\, dt \tag{25}$$

is the function usually measured in resonance experiments. Its real and imaginary parts χ' and χ'' may be easily obtained from $\Phi_{ij}(t)$. For the present example

$$\chi'_{11}(\omega) = \chi'_{22}(\omega) = \frac{1}{2[1 + (\lambda/S_0)^2]} \times \left[\frac{\omega_0 + \omega + \lambda/\tau}{(\omega_0 + \omega)^2 + (1/\tau)^2} + \frac{\omega_0 - + \lambda/\tau}{(\omega_0 - \omega)^2 + (1/\tau)^2} \right] \tag{26}$$

$$\chi''_{11}(\omega) = \chi''_{22}(\omega) = \frac{1}{2[1 + (\lambda/S_0)^2]} \times \left[\frac{\lambda(\omega_0 + \omega) - 1/\tau}{(\omega_0 + \omega)^2 + (1/\tau)^2} + \frac{\lambda(\omega - \omega_0) + 1/\tau}{(\omega_0 - \omega)^2 + (1/\tau)^2} \right] \tag{27}$$

In the limit $\lambda \to 0$, $\tau \to \infty$, thes results reduce to the well known expressions for ideal paramagnets, and for $\lambda = 0$ and $\tau = T_2$ they reproduce the results obtained from Bloch's equations.[11]

Case II. There are two minima of the interaction energy, namely for $\boldsymbol{\mu}$ parallel or antiparallel to the z-axis. Therefore,

$$P_{st}^{(0)}(\mathbf{x}^p) = \delta(x_p)\, \delta(y_p)\, \delta(z_p^2 - 1)$$

We obtain $\Phi_{12} = \Phi_{21} = \Phi_{13} = \Phi_{31} = \Phi_{23} = \Phi_{32} = \Phi_{33} = 0$ and $\Phi_{11}(t) = \Phi_{22}(t)$ given by Eq. (23), the same expression as for case I, but with $\tau = (\lambda^2 + S_0^2)/2Kv\lambda S_0$ and $\omega_0 = S_0/\lambda\tau$.

2.2. Numerical Simulations and Results

To calculate $\Phi_{ij}(t)$, Eq. (14), we must perform two ensemble averages, $\langle X_i(t \mid \mathbf{x}^p) \rangle_0$ and $\langle \cdots \rangle_{st}$. Since we do not have the corresponding probability densities $P^{(0)}(\mathbf{x}, t \mid \mathbf{x}^p, 0)$ and $P_{st}^{(0)}(\mathbf{x}^p)$, we use an alternative procedure, based on numerical simulations of the realizations of the stochastic process, governed by the Langevin equation in the absence of the perturbing field $F_i(t)$, i.e., keeping only the term $A_i^{(0)}$ of A_i. Equation (1) in discrete form reads[2]

$$X_i(t_n + \Delta t) = X_i(t_n) + A_i^{(0)}(\mathbf{X}(t_n))\, \Delta t + B_{ij}(\mathbf{X}(t_n))\, \sqrt{\Delta t}\, R_j(t_n) \tag{28}$$

where $R_j(t_n)$ are random numbers with normal distribution, $\langle R_j(t_n) \rangle = 0$, and $\langle R_i(t_n)\, R_j(t_{n'}) \rangle = \delta_{ij}\, \delta_{nn'}$.

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We begin by generating a stationary distribution of initial points $\mathbf{x}^p$, say NP of them. Each $\mathbf{x}^p$ may be obtained by starting from some arbitrary position in Γ, for example (1, 0, 0), and letting it evolve according to Eq. (28), for a sequence of $N \times M$ randomly chosen numbers $R_j(t_n)$ ($j = 1,..., M$ and $t_n = n\,\Delta t$, where $n = 0, 1, 2,..., N$), with the proper statistics and with N and NP sufficiently large so that the distribution of the $\mathbf{x}^p$ so obtained may be considered stationary. Each of the NP points $\mathbf{x}^p$ is then used as starting point for an ensemble of NR realizations of the stochastic process $\mathbf{X}$ driven again by Eq. (28). The arithmic average of the NR points $\mathbf{X}$ at time t is the expectation value $\langle \mathbf{X}(t \mid \mathbf{x}^p) \rangle_0$. Since we need for Eq. (14) the partial derivatives of this expectation value with respect to the initial components x_k^p, $k = 1,..., M$, we have to repeat the NR realizations M times, starting each time from a different point $\mathbf{x}^p + \Delta x_k \hat{e}_k$ very near to $\mathbf{x}^p$, (i.e., $\Delta x_k \ll 1$),

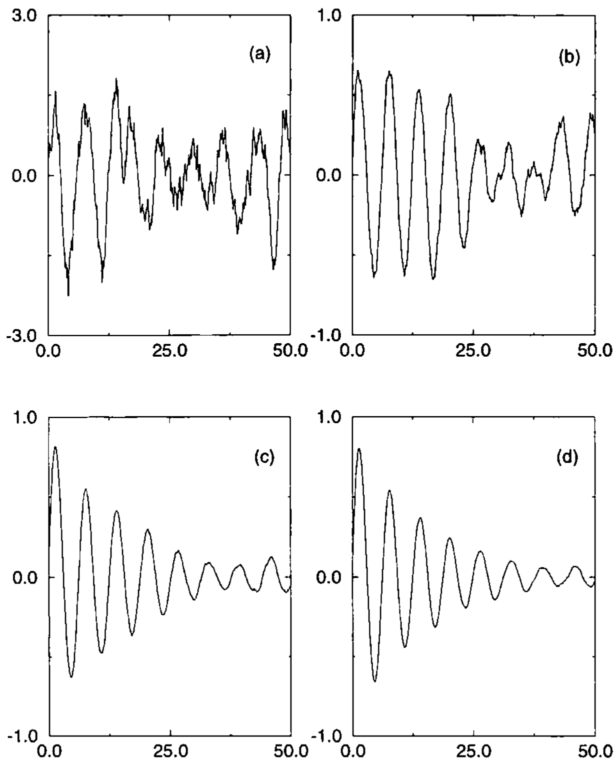

Fig. 1. Influence of the size of the ensemble on the calculation of the averages in Eq. (14) for $\Phi_{11}(t)$ versus time for case I. (a–d) $NP = NR = 8, 64, 256$, and 512, respectively. The parameters are $Q = S_0 H_{\rm eff}/k_B T = 1.0$, $\bar{\lambda} = 0.1$, $\bar{\beta} = 1.0$, $\bar{D} = 0.01$.

where $\hat{e}_k$ is the unit vector in the k-direction. The partial derivative is then calculated as

$$\frac{\partial}{\partial x_k^p}\langle X_i(t\,|\,\mathbf{x}^p)\rangle_0 \simeq \frac{\langle X_i(t\,|\,\mathbf{x}^p+\Delta x_k\hat{e}_k)\rangle_0-\langle X_i(t\,|\,\mathbf{x}^p)\rangle_0}{\Delta x_k} \tag{29}$$

The arithmetic average over the NP points $\mathbf{x}^p$ of the product of this derivative by γ_{kj} summed over k is the response function $\Phi_{ij}(t)$ as given by Eq. (14). The complex admittance (25) may be obtained by computer help of the fast Fourier transform (FFT) routine.

Using the superconductor CRAY Y-MP2E of our university, we made simulations for cases I and II considered above with several distinct values of NP, NR, and Δt. In all simulations the values $\Delta x_p = \Delta y_p = \Delta z_p = 0.05$

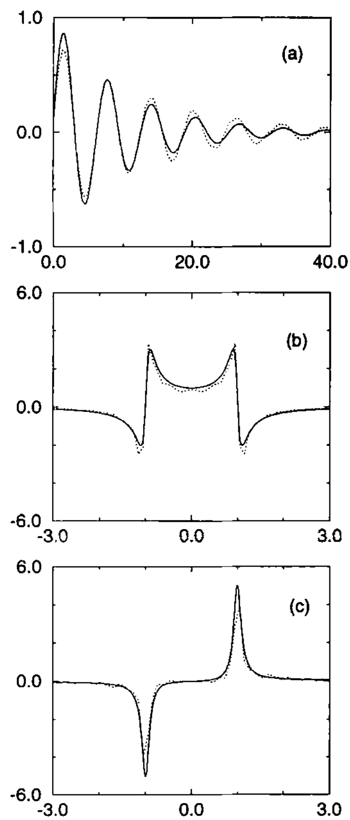

Fig. 2. (a) $\Phi_{11}(t)$ versus time, (b) χ' versus ω, and (c) χ'' versus ω for case I, with $Q = 1.0$, $\bar{\lambda} = 0.1$, $\bar{\beta} = 1.0$; solid line: $\bar{D} = 0$; dotted line: $\bar{D} = 0.02$.

were used. To obtain the normally distributed random numbers R_j we used the ranf($\cdot$) function of the CRAY, which generates pseudo-random numbers uniformly distributed between 0 and 1 in a fully vectorized way and with a period equal to $2^{46} \approx 10^{13}$. The normally distributed numbers R_j were then obtained by the polar method.[12] The enormous period of ranf($\cdot$) guarantees that the sequence of generated random numbers does not close it itself during a typical simulation. For example, for a rather rigorous simulation, using $NP = NR = NT = 1000$, where NT is the number of steps of integration, we need $3 \times 4 \times NP \times NR \times NT = 1.2 \times 10^{10}$ random numbers, still far from ranf($\cdot$)'s period. To put Eq. (28) into a form completely dimensionless appropriate for numerical simulations, we used the following dimensionless quantities: $\bar{t} = (k_B T/S_0) \times t$, $\bar{V} = V/k_B T$, $\bar{\lambda} = \lambda/S_0$, $\bar{\beta} = \beta S_0/k_B T$, $\bar{D} = D/S_0 k_B T$, and $\mathbf{H} = -\partial\bar{V}/\partial\mathbf{X}$.

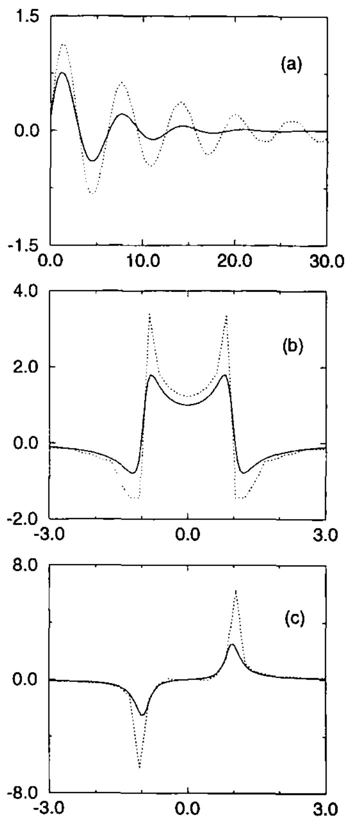

Fig. 3. Same as Fig. 2, but for $\bar{\beta} = 0.1$.

To get a feeling for the effect of the size of the ensemble used for calculating the averages involved in Eq. (14), we show in Fig. 1 a sequence of results for Φ_{11} calculated for case I (external field), with different numbers of particles and realizations, keeping $NP = NR$. Figure 1a shows the result of Eq. (14) obtained for a very poor averaging procedure, with only eight particles and eight realizations of the SP for each particle, i.e., $NP = NR = 8$. Figures 1b–1d show the ensemble averages for $NP = NR = 64$, 256, and 512, respectively. It can be seen that the last curve corresponds to rather good statistics.

Figures 2 and 3 are also results of the simulations for case I, varying the parameters $\bar{\beta}$ (relaxation constant of longitudinal fluctuations) and $\bar{D}$ (noise intensity). The zero-noise limit, $\bar{D} = 0$, is shown in all figures by the solid lines. In Fig. 2 the dotted lines show the results for $\bar{\beta} = 1$ and

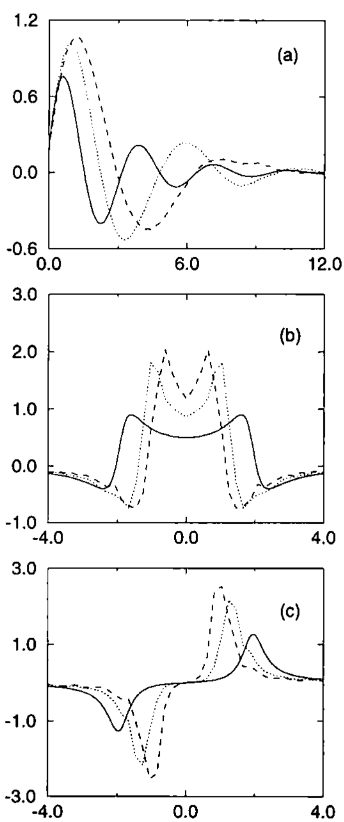

Fig. 4. Same as Fig. 2, but for case II, $Q \equiv Kv/k_B T$, $\bar{\lambda} = 0.2$, and $\bar{\beta} = 0.1$; solid line: $\bar{D} = 0$, dotted line: $\bar{D} = 0.01$; dashed line: $\bar{D} = 0.02$.

$\bar{D}=0.02$. With these values of the parameters there are no important longitudinal fluctuations and our model reduces to Brown's model.[9] In Fig. 3 the dotted line is for $\bar{\beta}=0.1$ and $\bar{D}=0.02$. Important fluctuations in the magnitude of $\boldsymbol{\mu}$ occur for these values of the parameters. We see clearly that the noise enhances the response of the system to the transverse field. The influence of the noise on the response of a system to a deterministic signal, enhancing the resonance intensity, is a remarkable result. Similar phenomena, referred to as *stochastic resonance*, have been reported in the literature (e.g., ref. 13) and have aroused considerable interest during the last few years. Our results show that also in superparamagnetic systems stochastic resonance can be found as long as the particles are so small that important fluctuations in the magnitude of the magnetic moment should be expected. This is not the first time that stochastic resonance has been predicted theoretically in superparamagnets, but the present model, method of solution and results are very different from those of previous works.[14,15] In the Brown model limit, Fig. 2, our results do not show evidence for stochastic resonance; in this case the noise has the effect of attenuating the resonance intensity, as we see in Fig. 2c.

Figure 4 shows the results for case II, for $\bar{\beta}=0.1$ and $\bar{D}=0$ (solid line), $\bar{D}=0.01$ (dotted line) and $\bar{D}=0.02$ (dashed line). In this case one sees that the noise not only enhances the response of the system, but also decreases the resonance frequency.

APPENDIX A

Let us formulate the detailed balance conditions for the FPE in the usual way:[4]

Given that, under time-reversal transformation, $\mathbf{x}\rightarrow\varepsilon\mathbf{x}=(\varepsilon_1 x_1, \varepsilon_2 x_2,..., \varepsilon_M x_M)$, with $\varepsilon_i=\pm 1$, then

$$\varepsilon_i A_i^{(0)}(\varepsilon\mathbf{x})\, P_{st}^{(0)}(\mathbf{x}) = -A_i^{(0)}(\mathbf{x})\, P_{st}^{(0)}(\mathbf{x}) + \sum_{j=1}^{M} \partial_j[D_{ij}(\mathbf{x})\, P_{st}^{(0)}(\mathbf{x})] \tag{A1}$$

and

$$\varepsilon_i\varepsilon_j D_{ij}(\varepsilon\mathbf{x}) = D_{ij}(\mathbf{x}) \tag{A2}$$

Since under time-reversal $\boldsymbol{\mu}\rightarrow-\boldsymbol{\mu}$ and therefore $\mathbf{x}\rightarrow-\mathbf{x}$, we have $\varepsilon_1=\varepsilon_2-\varepsilon_3=-1$. Then the condition (A1) can be written in the form

$$\tilde{A}_i^{(0)}(\varepsilon\mathbf{x})\, P_{st}^{(0)}(\mathbf{x}) = \tfrac{1}{2}\sum_{j=1}^{3} \partial_j[D_{ij}(\mathbf{x})\, P_{st}^{(0)}(\mathbf{x})]$$

where $\tilde{A}_i^{(0)}(\mathbf{x}) \equiv \frac{1}{2}[A_i^{(0)}(\mathbf{x}) - A_i^{(0)}(-\mathbf{x})]$. As the diffusion matrix in this cas is not singular, $D_{ij}^{-1} = \delta_{ij} S_0^2/2D$, and we can invert the above equation to obtain

$$\partial_k[\ln P_{st}^{(0)}(\mathbf{x})] = \frac{S_0^2}{D} \tilde{A}_i^{(0)}(\mathbf{x})$$

But it is easy to verify that $\nabla \times \tilde{\mathbf{A}} \neq 0$ for both cases in Section 3. Therefore this equation cannot be satisfied, since ist left-hand side is a gradient, and consequently Eq. (A1) is also not satisfied.

APPENDIX B

In the absence of noise ($D = 0$) and knowing $A_i^{(0)}(\mathbf{x})$ we can integrate Eq. (1). We obtain for case I (uniform static magnetic field)

$$z(t \mid \mathbf{x}^p) = \frac{\sinh(t/\tau) + z_p \cosh(t/\tau)}{\cosh(t/\tau) + z_p \sinh(t/\tau)}$$

$$x(t \mid \mathbf{x}^p) = \frac{\sqrt{1 - z_p^2} \cos(\omega_0 t - \phi_p)}{\cosh(t/\tau) + z_p \sinh(t/\tau)}$$

and

$$y(t \mid \mathbf{x}^p) = -\frac{\sqrt{1 - z_p^2} \sin(\omega_0 t - \phi_p)}{\cosh(t/\tau) + z_p \sinh(t/\tau)}$$

where

$$\tau = \frac{S_0^2 + \lambda^2}{\lambda H^{(0)} S_0}$$

and

$$\omega_0 = \frac{S_0}{\lambda \tau}$$

For the case II (uniaxial crystal field) we obtain

$$z(t \mid \mathbf{x}^p) = \frac{z_p}{[z_p^2 + (1 - z_p^2) \exp(-2t/\tau)]^{1/2}}$$

$$x(t \mid \mathbf{x}^p) = \frac{\exp(-t/\tau)}{[z_p^2 + (1 - z_p^2) \exp(-2t/\tau)]^{1/2}} \times [x_p \cos \Omega(t, z_p) + y_p \sin \Omega(t, z_p)]$$

and

$$y(t\,|\,\mathbf{x}^p) = \frac{\exp(-t/\tau)}{[z_p^2 + (1-z_p^2)\exp(-2t/\tau)]^{1/2}} \times [y_p \cos\Omega(t, z_p) + x_p \sin\Omega(t, z_p)]$$

where

$$\Omega(t, z_p) = \omega_0 z_p \int_0^t \frac{dt'}{[z_p^2 + (1-z_p^2)\exp(-2t'/\tau)]^{1/2}} = \frac{S_0}{\lambda} \ln\left\{\frac{[1 + z_p^2(e^{2t/\tau} - 1)]^{1/2} + z_p e^{t/\tau}}{1 + z_p}\right\}$$

$$\tau = \frac{S_0^2 + \lambda^2}{2Kv\lambda S_0}$$

$$\omega_0 = \frac{S_0}{\lambda\tau}$$

Observe that $\Omega(0, z_p) = 0$, $\Omega(t, \pm 1) = \pm\omega_0 t$, and

$$\lim_{t\to\infty} = \omega_0 t + \ln\left(\frac{2z_p}{1+z_p}\right)^{S_0/\lambda}$$

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